

**REMARKS**

The Official Action dated May 15, 2009 has been carefully considered. Accordingly, the present Amendment is believed sufficient to place the present application in condition for allowance. Reconsideration is respectfully requested.

By the present amendment, claims 3 and 4 are cancelled. Claim 5 is amended to provide antecedent basis for recitation of “the system” and for several matters of form. Claim 5 is also amended to recite the upper limit of the proportion of alkali metal hydroxide to alkali metal hydrosulfide as 1.02 and the upper limit of the ratio of alkali metal hydroxide to charged sulfur source as 1.050, both in accordance with Example 1 in the present specification (see Table 1 at page 38). Claim 5 is also amended to clarify that the discharge of hydrogen sulfide to the exterior of the system is directly linked with a weight loss of sulfur amount in the system, as described at page 17, lines 15-17 of the specification, and to define the [Charged sulfur source] as [Total moles of sulfur charged] – [Moles of sulfur volatized out after dehydration] in accordance with the specification at page 18, lines 11-14. Finally, claims 17 and 18 are added, support for which is found in Example 2 and Table 2 at page 39 of the specification. It is believed that these changes are fully supported by the original specification, whereby entry of the present Amendment is in order and is respectfully requested.

In the Official Action, claim 5 was objected to as containing a redundant recitation of the proportion of alkali metal hydroxide to alkali metal hydrosulfide. Claim 5 as amended omits a second recitation of the proportion. Accordingly, the objection has been overcome and reconsideration is respectfully requested.

Claims 3, 5, 7 and 10-13 were rejected under 35 U.S.C. 102(b) as being unpatentable over Miyahara et al, US 5,840,830. The Examiner asserted that Miyahara et al teach that sodium sulfide can be produced by in situ reaction of sodium hydrosulfide and sodium hydroxide

(column 4, lines 61-64), that these reactants can be used in equimolar amounts (column 6, lines 49-53), and that, if such a step is employed, the resulting ratio of NaOH to S is (23.45/22.83) or 1.027 (page 5), whereby Miyahara et al anticipate the process of claim 5. The Examiner further asserted that if the sodium sulfide of Miyahara et al is prepared in situ by the reaction of an almost equimolar amount of alkali metal hydrosulfide and alkali metal hydroxide, the reaction inherently produces the alkali metal sulfide and water as an aqueous mixture. The Examiner also asserted that Miyahara et al explicitly teach that water or the azeotropic mixture of water and the organic amide solvent is discharged out of the system along with hydrogen sulfide at column 7, lines 44-48, and there is nothing in the Applicants' claims which excludes the recovery and reuse of the hydrogen sulfide.

This rejection is traversed and reconsideration is respectfully requested. Applicants submit that Miyahara et al do not teach the process of claim 5 or the improvements thereof in providing poly(arylene sulfide) (PAS) with an improved combination of properties. More particularly, the process for producing a PAS according to the present invention and as defined in claim 5 comprises four important process steps which, in combination, provide a PAS having a desirable combination of improved properties as also set forth in claim 5. Specifically, in the dehydration step (1), an organic amide solvent, and an alkali metal hydrosulfide and an alkali metal hydroxide, both as aqueous mixtures, are heated and reacted in a proportion of 0.95 to 1.02 mol of alkali metal hydroxide per mol of the alkali metal hydrosulfide, wherein distillate containing water is discharged to the exterior of the system and hydrogen sulfide formed upon the dehydration is discharged as a gas to the exterior of the system. As the discharge of hydrogen sulfide to the exterior of the system is directly linked with a weight loss of sulfur amount in the system, it is evident that the discharged hydrogen sulfide is not recycled into the reaction system.

In the subsequent charging step (2), the total number of mols of (i) alkali metal hydroxide formed with hydrogen sulfide upon the dehydration, (ii) the alkali metal hydroxide added prior to the dehydration, and (iii) the alkali metal hydroxide added after the dehydration is 1.015 to 1.050 mols per mol of the charged sulfur source and the mols of water are controlled to provide 0.5 to 2.0 mols of water per mol of the charged sulfur source. The amount of the charged sulfur source is calculated out in accordance with the equation: [Charged sulfur source] = [Total moles of sulfur charged] – [Moles of sulfur volatilized out after dehydration], again evidencing that the discharged hydrogen sulfide is not recycled into the reaction system. Additionally, the polymerization is conducted in the defined first-stage polymerization step (3) and the defined second-stage polymerization step (4) to provide a polymer as claimed.

The combination of the defined steps (1)-(4) is important in providing a PAS polymer product in high yield and having good melt viscosity, low yellowness index, low dimer byproduct, good reactivity with aminosilane coupling agents, and low volatility from the alkali metal hydrosulfide and alkali metal hydroxide reactants. It is preferred to use the claimed alkali metal hydrosulfide and an alkali metal hydroxide as starting reactants as they are less expensive than the traditionally employed alkali metal sulfide. However, in the past, it has been difficult to produce a polymer having a good combination of properties in a stable manner using these reactants, as described in the present specification, beginning at page 2, line 17. As further described in the present specification, for example beginning at page 17, line 6, the alkali metal hydroxide reacts with the organic amide solvent in the dehydration heat treatment to form an alkali metal alkylaminoalkanoate, and the alkali metal hydrosulfide exists in the system in the form of a complex with the alkali metal alkylaminoalkanoate. On the other hand, a part of the alkali metal hydrosulfide reacts with water to form hydrogen sulfide and an alkali metal hydroxide, and the hydrogen sulfide formed is discharged outside the system. The steps recited

in claim 5 control these various reactions arising from the use of the alkali metal hydrosulfide and alkali metal hydroxide reactants to provide a stable reaction and a polymer product having a consistently good combination of properties as recited.

Miyahara et al disclose a process for producing poly(arylene sulfide) wherein an alkali metal sulfide such as sodium sulfide is reacted with a dihaloaromatic compound, and wherein hydrogen sulfide vaporized off during the dehydration step is recovered and reused. Miyahara et al indicate that the sodium sulfide can be produced by in situ reaction of sodium hydrosulfide and sodium hydroxide (column 4, lines 61-64) and that these reactants can be used in equimolar amounts (column 6, lines 49-53). Importantly, Miyahara et al fail to exemplify any process wherein sodium sulfide is formed in situ as required by present claim 5.

Miyahara et al also fail to exemplify a dehydration step as recited in claim 5, wherein a mixture containing (i) organic amide solvent, (ii) an alkali metal hydrosulfide as an aqueous mixture with water, and (iii) an alkali metal hydroxide as an aqueous mixture with water, are heated to discharge at least a part of a distillate containing water from the interior of the system containing the mixture to the exterior of the system and to discharge hydrogen sulfide formed upon the dehydration as a gas to the exterior of the system. To the contrary, Miyahara et al specifically describe the use of sodium sulfide as a reactant and require that the hydrogen sulfide formed in the dehydration is recovered and reused.

The Examiner asserted in the Official Action that the present claims do not exclude the recovery and reuse of the discharged hydrogen sulfide. Applicants submit however that the present claim 5, in reciting discharge of at least a part of a distillate containing water from the interior of the system to the exterior of the system, wherein hydrogen sulfide formed upon the dehydration is discharged as a gas to the exterior of the system, in specifying that the discharge of hydrogen sulfide to the exterior of the system is directly linked with a weight loss of sulfur in

the system, and in defining the charged sulfur source as the total moles of sulfur charged less the moles of sulfur volatized out after dehydration, preclude the recovery and reuse taught by Miyahara et al. In contrast, Miyahara et al disclose that hydrogen sulfide is discharged together with water or an azeotropic mixture of water and organic amide solvent and subsequently recovered (see, for example, column 7, lines 44-55 and the Examples), whereby the hydrogen sulfide is maintained in the reaction system according to Miyahara et al.

These are critical differences between the teachings of Miyahara et al and the process of the present invention as Applicants have discovered that the in situ formation of sodium sulfide from an alkali metal hydrosulfide and an alkali metal hydroxide in the dehydration reaction requires careful control to reliably provide a PAS having a desirable combination of properties. Miyahara et al do not teach the careful control that is necessary and, in fact, the processes exemplified by Miyahara et al, if modified to include the in situ formation of sodium sulfide, do not result in the process of claim 5. More specifically, in Example 1a of Miyahara et al, which recycles discharged hydrogen sulfide, 30 g of 97% NaOH was first added to the dehydration system (column 12, lines 16-19). Miyahara et al disclose that when an alkali metal hydroxide exists in the reaction system, hydrogen sulfide is converted into its corresponding alkali metal sulfide (column 8, lines 34-36). Thus, if the process of Example 1a of Miyahara et al uses an alkali metal hydrosulfide and an alkali metal hydroxide in equimolar amounts, the additional alkali metal hydroxide is required for reused hydrogen sulfide, in addition to the alkali metal hydroxide used in an equimolar amount as the alkali metal hydrosulfide. In short, according to Miyahara et al, the alkali metal hydroxide in an equimolar amount to the alkali metal hydrosulfide and the alkali metal hydroxide for the reused hydrogen sulfide are both required.

For example, in Example 1a of Miyahara et al, the alkali metal hydrosulfide and the alkali metal hydroxide in equimolar amounts would be 22.49 mol of each. However, the

additional 30 g of 97% NaOH taught by Miyahara et al, comprising 0.72 mol NaOH, results in a NaOH/NaSH ratio of  $(22.49 + 0.72) / 22.49$ , which is equal to 1.032. This ratio is outside the NaOH/NaSH range of 0.95 to 1.02 required in the process of claim 5. Examples 1b, 2 and 5 of Miyahara et al are similar to Example 1a. In Example 3a, the equimolar amounts of alkali metal hydrosulfide and alkali metal hydroxide would be 21.28 mol each. Miyahara et al disclose the addition of 30 g of 97% NaOH (0.72 mol NaOH), resulting in an NaOH/NaSH ratio of  $(21.28 + 0.72) / 21.28$ , which is equal to 1.033. Example 3b of Miyahara et al is similar to Example 3a. In Example 4a, the equimolar amounts of alkali metal hydrosulfide and alkali metal hydroxide would be 21.28 mol each. Miyahara et al disclose the addition of 50 g of 97% NaOH (1.21 mol), resulting in an NaOH/NaSH ratio of  $(21.28 + 1.21) / 21.28$ , which is equal to 1.056. Example 4b of Miyahara et al is similar to Example 4a. Thus, according to the teachings of Miyahara et al, the discharged hydrogen sulfide is recycled and the NaOH/NaSH ratio is above the upper limit of 1.02 required in the process of claim 5.

On the other hand, the present specification shows that when the NaOH/NaSH ratio is above 1.02, the properties of the resulting PAS begin to decline. More specifically, in Example 3 in the specification, wherein the NaOH/NaSH ratio is 1.04 before dehydration (comparable to that of Example 1a of Miyahara et al), and the ratio of NaOH to charged sulfur source is 1.075, the PAS yield is lowered, as is the melt viscosity of the PAS, while the yellow index and the amount of dimer impurity are both increased. Thus, not only does the modification of the Miyahara et al process to include in situ sodium sulfide formation not meet the process requirements of claim 5, the resulting PAS exhibits a less desirable combination of properties.

Finally, Applicants note that the Comparative Examples of Miyahara et al do not employ hydrogen sulfide recycle or additional NaOH. However, Miyahara et al do not provide any

teaching or suggestion that an equimolar amount of NaOH and NaSH should be used in such a process, particularly to result in improved properties in the thus formed PAS.

Anticipation under 35 U.S.C. §102 requires that each and every element as set forth in the claims is found, either expressly or inherently described, in a single prior art reference. *In re Robertson*, 169 F.3d 743, 745, 49 U.S.P.Q. 2d 1949, 1950 (Fed. Cir. 1999). Miyahara et al fail to teach a PAS production method wherein sodium sulfide is formed in situ in a dehydration step using an NaOH/NaSH ratio of 0.95 to 1.02, particularly in the absence of hydrogen sulfide gas recycling. Thus, Miyahara et al do not expressly or inherently describe each and every element as set forth in claim 5. Accordingly, Miyahara et al do not anticipate claim 5, or any of claims 7 and 10-13 dependant on claim 5, under 35 U.S.C. §102. The rejection has therefore been overcome and reconsideration is respectfully requested.

Finally, in the Official Action, the Examiner withdrew the rejection based on Sato et al, JP 2000-191785. Sato et al, like Miyahara et al, generally teach that sodium sulfide can be produced by in situ reaction of sodium hydrosulfide and sodium hydroxide in paragraph [0018] and that these reactants can be used in equimolar amounts in paragraph [0026]. Importantly, however, Sato et al do not exemplify any process combining use of the alkali metal hydrosulfide and the alkali metal hydroxide as the sulfur source, particularly in accordance with steps (1)-(4) as set forth in claim 5. Moreover, even if one of ordinary skill in the art were to employ an in situ sodium sulfide formation in the exemplified method of Sato et al's Example 1, a process as presently claimed does not result.

Particularly, Sato et al disclose in Example 1 that 13.3 g of NaOH having a purity of 97% was added in such a manner that the total NaOH in the reactor was 6.00 mol% to the available Na<sub>2</sub>S (paragraph [0057]). The “6.00 mol%” in this description is understood to mean that the total NaOH in the reactor is increased by 6.00 mol% to the available Na<sub>2</sub>S and, accordingly, it is

understood that Sato et al disclose that, if the equimolar amounts of NaOH and NaSH are employed for sodium sulfide formation, the total NaOH in the reactor/available Na<sub>2</sub>S is 1.060. A ratio of NaOH/available S of 1.060 is outside the range of 1.015 to 1.050 in the charging step of present claim 5. Moreover, Example 3 in the present specification shows that as the NaOH/available S ratio is increased above 1.050 in the reactor before initiation of the first-stage polymerization step, particularly with an NaOH/NaSH ratio of 1.04, the properties of PAS begin to deteriorate in yield, melt viscosity, color tone (yellow index), content of the dimer impurity, and reactivity to aminosilane. Thus, Sato et al similarly fail to disclose the claimed process and the improvements in the PAS provided thereby.

It is believed that the above represents a complete response to the Official Action and places the present application in condition for allowance. Reconsideration and an early allowance are requested.

Please charge any fees required in connection with the present communication, or credit any overpayment, to Deposit Account No. 503915.

Respectfully submitted,

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